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Abstract
This paper examines the reliability of a widely used method for temperature determination by multi-wavelength pyrometry. In recent WDM experiments with ion-beam heated metal foils, we found that the statistical quality of the fit to the measured data is not necessarily a measure of the accuracy of the inferred temperature. We found a specific example where a second-best fit leads to a more realistic temperature value. The physics issue is the wavelength-dependent emissivity of the hot surface. We discuss improvements of the multi-frequency pyrometry technique, which will give a more reliable determination of the temperature from emission data.

Keywords: warm dense matter, temperature measurement, pyrometry, heavy ion beam heating.

Introduction

Warm dense matter can be produced by several heating technologies including lasers, pulsed electric current, electron, ion and x-ray beam heating, shock-wave heating, etc (DOE, 2009; Hoffmann *et al*. 2005; Sasaki *et al*. 2006; Ng *et al*. 2005; Lee *et al*. 1998). Temperature measurements are essential to WDM science. In most cases the deposited energy is determined accurately but at present there is no universal method to directly measure the temperature. As a consequence, the specific heat $C = dE/dT$ is not accurately determined and this is a key unknown aspect of the equation of state (EOS). There is also a class of experiments, which aims to study phase transitions: these may be high-temperature evaporation of refractory metals, a liquid-vapor critical point, or an abrupt change of electronic structure (metal-insulator transition). Such transitions typically occur at definite temperatures and an accurate measurement of the transition temperature is obviously a key step for building up the quantitative science of warm dense matter. Today the critical points of refractory metals are uncertain to something like 50 % and this uncertainty is a leading source of EOS uncertainty for WDM expanded below solid density. At higher energy-density (HEDP), shock Hugoniot measurements can accurately determine for the material density, energy-density and pressure, but different EOS models predict different temperatures for the same (energy/pressure/density) shocked conditions, so again temperature is a key uncertainty for HEDP EOS data. Finally, almost all the computer codes used to predict the properties of warm dense matter (or hotter plasmas) use density and temperature as inputs and for this reason it will be easier to directly compare theory and experiment if one can accurately measure the temperatures attained in any given experiment.

Pyrometry (Dewitt, 1998), the determination of surface temperature by analysis of thermally emitted light, is a well-known and commonly used method for measuring temperature of hot matter. While being a popular technique, pyrometry is known to two drawbacks: a limitation to measure only the surface temperature, and limited accuracy in experiments where surface emissivity is not known. This paper will discuss the second issue.

The normal surface emissivity $E(\lambda)$ is (by definition) the ratio of the observed emission intensity to the black-body function $B_\lambda(T)$:

$$I(\lambda) = E(\lambda) * B_\lambda(T)$$

The difficult point is that while the surface emissivity is known for cold matter (tabular data exists), it is generally unknown above melting ($T \geq$ 2000 K-3000 K) temperatures, i.e. in the WDM regime. The fundamental reason for this is the fact the high-frequency (optical) AC conductivity, or equivalently the optical constants n, k, which determine the emissivity, are not known for material at WDM conditions.

An attempt to improve the situation by simultaneously measuring emission at many wavelengths (multi-wavelength pyrometery)(Dewitt, 1998; Ni *et al*. 2008; Ni *et al*. 2011) does not solve the problem because there is a new unknown emissivity $E(\lambda)$ at each new wavelength λ and there always N+1 unknowns, where N is the number of direct measurements. Thus from the mathematical point of view, multi-wavelength pyrometry cannot produce a unique temperature. If emissivity is not measured and there is no information about its behavior, experimenters can assume a linear or quadratic dependence of $E(\lambda)$ on wavelength λ and optimize this "gray-body" fit to the multi-frequency emission measurements. The temperature produced by this fit is often reported as the temperature "observed" in the experiment.

Real-world example of "blind" curve fitting.

Unfortunately the accuracy of temperature determination does not reflect the accuracy of gray-body fits to spectral data. A recent real-life example, which motivates this paper, was encountered during WDM experiments at the NDCX-I, heavy ion induction LINAC (Bieniosek *et al*. 2010). In the experiments, a gold foil of thickness 0.15 μm was heated by a 300 keV K^+, 250 kW/cm^2 beam and the foil temperature was measured by a standard high-speed streak-camera optical pyrometer (SOP) technique (Bieniosek2 *et al*. 2010): thermal emission was recorded by a system consisting of a spectral grating and a fast optical streak camera. The entire SOP setup was calibrated absolutely with a tungsten ribbon lamp, which is a NIST-traceable calibration source. Absolute emission spectra from 400 nm to 850 nm were recorded with ~2% accuracy and a time-resolution of approximately 50 nsec (Figure 1). The data were analyzed by the usual method of optical pyrometry, i.e the emission spectrum was fit to a blackbody function multiplied by an emissivity $E(\lambda)$, which was assumed to be a polynomial (in our case quadratic) function of a wavelength in the visible range.

The resulting best fit, shown in Figure 2, leads to a temperature of 7200 K (the spectrum shown corresponds to a time of 800 ns, where the radiation is a maximum). The best least-square fit is determined by the global minimum of the sums of residuals. The deviation of the fit from the experimental data is very small, and if one had stopped here one would have believed the temperature.

However the energy deposited by the NDCX-I beam is well known (fluence is ~250 kW/cm^2), and with all the energy accounted for, the highest expected temperature could not exceed 3400 K (Bieniosek *et al*. 2010). The energy required to heat a 0.150 micron gold foil up to the boiling point (not counting the latent heat of evaporation) is approximately 0.2 J/cm^2. To deliver this energy in 1 μsec requires already 200 kW/cm^2. A thick target would be cooled by thermal conduction, but our thin targets do not cool in this way (lateral thermal conduction is negligible on our time-scale). However, the target cools by evaporation at high temperatures. We estimate the cooling rate at a temperature equal to the normal boiling point, T_b = 3240 K for Au. At this temperature the saturation pressure of gold vapor would be 1 atmosphere. This corresponds to a gas density of approximately 2.24×10^{18} $atoms/cm^3$. In equilibrium the evaporation rate would just balance condensation from the saturated vapor, but in vacuum there is little condensation. Assuming a sticking coefficient close to unity, that balance implies an evaporation rate of $3.3\ 10^{22}$ $atom/cm^2/sec$. Each evaporated atom carries away an energy of approximately the latent heat of evaporation, ~ 3.51 eV for gold. Thus the evaporation cooling heat flux is ~ 18.5 kW/cm^2. This should be multiplied by a factor 2 for a thin foil or a factor 4 for spherical droplets (ratio of evaporating surface area to projected area facing the beam). If the ion heating does not deposit more energy than 270 kW/cm^2 (= 200 kW/cm^2 for initial temperature rise + 70 kW/cm^2 for evaporative cooling), the target is expected to reach a temperature lower than the normal boiling point of 3240 K. Based on these numbers, a temperature of 7200 K is not possible with the NDCX-I ion beam.

After a careful analysis triggered by this discrepancy, a "second-best" optimum fit to the same data, which yields a much lower temperature, 2400 K (Figure 3) was found. Both Newton-Gauss and Levenberg-Marquardt, algorithms commonly used least-square optimization algorithms (Ni *et al*. 2008;), lead to the same results. The 2400 K fit is in much better agreement with the thermodynamic analysis but is not such a nice fit to the measured spectrum from a statistical point of view. However, it is important to note that the emissivity $E(\lambda)$ resulting from the 2400 K fit (Figure 3b) is very much like the emissivity of normal Au in this spectral range (Watanabe *et al*. 2003), while the emissivity inferred from the high-temperature fit in Figure 2b has the opposite curvature.

The summary of the comparison is that the beautiful fit in Figure 1a is a 7200 K blackbody source times the emissivity of Fig. 2b. The less beautiful fit in Figure 3a is a 2400 K blackbody times the emissivity of Figure 3b. But for two good reasons - the total energy available and the shape of Figure 3b - the second fit is much more likely to be correct. The important lesson is that, generally speaking, *the accuracy of temperature determination has no relation to the accuracy of the fit to spectral data*. One cannot measure the accuracy of a temperature extracted from conventional pyrometry by the

quality of the curve-fit to the spectrum: looking only at Figure 1a, it would look like 1% precision, but seeing the whole picture, the temperature error from Fig. 1a is ~ 50 %!

Polarization of self emission as a key to solve the ambiguity problem

An approach to address the temperature ambiguity, inherent to multi-wavelength pyrometers, is to find other manifestations of emissivity. It turns out that in addition to self-emission spectra, there is more information available which is generally overlooked, namely the polarization of the self-emission, occurring at a non-zero angle to a surface: generally s-polarized (parallel to surface) E-field emission is less intense than p-polarized emission. If a surface is perfectly smooth and clean, and the material beneath the surface is homogenous, the reflectivity, r =1-ε is related by Fresnel to the complex dielectric function, ξ (λ). For a non-zero angle from the surface normal, α, the reflectivity r is different for the *p* and *s* components of electrical vector, and the polarized spectral emission density can be described by

$$I_p(\lambda,\alpha) = \left(1 - \left|\frac{\xi(n,k)\cdot\cos(\alpha) - \sqrt{\xi(n,k) - \sin^2(\alpha)}}{\xi\cdot\cos(\alpha) + \sqrt{\xi - \sin^2(\alpha)}}\right|^2\right) * B_\lambda(T),$$

$$I_s(\lambda,\alpha) = \left(1 - \left|\frac{\cos(\alpha) - \sqrt{\xi(n,k) - \sin^2(\alpha)}}{\cos(\alpha) + \sqrt{\xi(n,k) - \sin^2(\alpha)}}\right|^2\right) * B_\lambda(T),$$

$$\xi = (n - ik)^2 - \text{complex dielectric function.}$$

Equation 2: Spectral emission density of p- and s-polarization components at angle α and wavelength, λ.

As one can see, if the absolute intensities of polarization components at (at least) two angles (four values in total) are measured, there is a *unique* solution for T, and optical constants, n and k. One can take data at more angles and obtain an over-determined system of equations, which is favorable when measurements have finite accuracy. In contrast to the multi-wavelength pyrometery, addition of more measurements at different wavelengths does not improve accuracy since there are always more unknowns than measured values. The combination of polarization and multi-wavelength pyrometer data, can thus reduce the overall ambiguity of temperature determination. A diagnostic based on the above principle, is called *polarization pyrometer* and is described in details in (More *et al*. 2010; Ni *et al*. 2012).

Conclusion

The weak point of the pyrometry technique is the fact that, over the visible range, blackbody functions with different temperatures (combined with different emissivities) can fit the emission data almost equally well. The example in Figures 1-4 utilizes precisely the usual procedure in optical pyrometry and very similar methods are used in many laboratories for determining temperatures produced by shock-waves or other warm-

dense matter experiments. A practical consequence from this finding is that if possible, emssivity should be measured independently and common sense verification of maximum temperature must be carried out. We were lucky to have a reference to emissivity value, but what about the experiments at much higher temperatures, where there is no emissivity or beam fluence reference? Addition of polarization resolved measurements to the spectral emission has a potential to solve the ambiguity problem.

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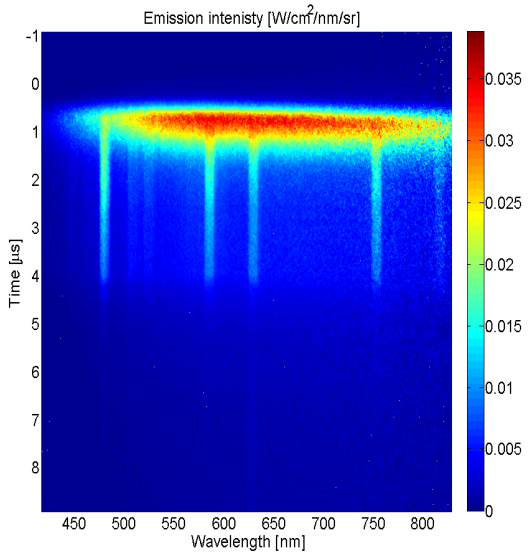

Figure 1: Absolute emission spectrum as a function of time from a beam heated Au target by NDCX-I. Temperature is determined from spectrum at each moment of time. Lines corresponds to collisional Au-I vapor excitation by ion beam.

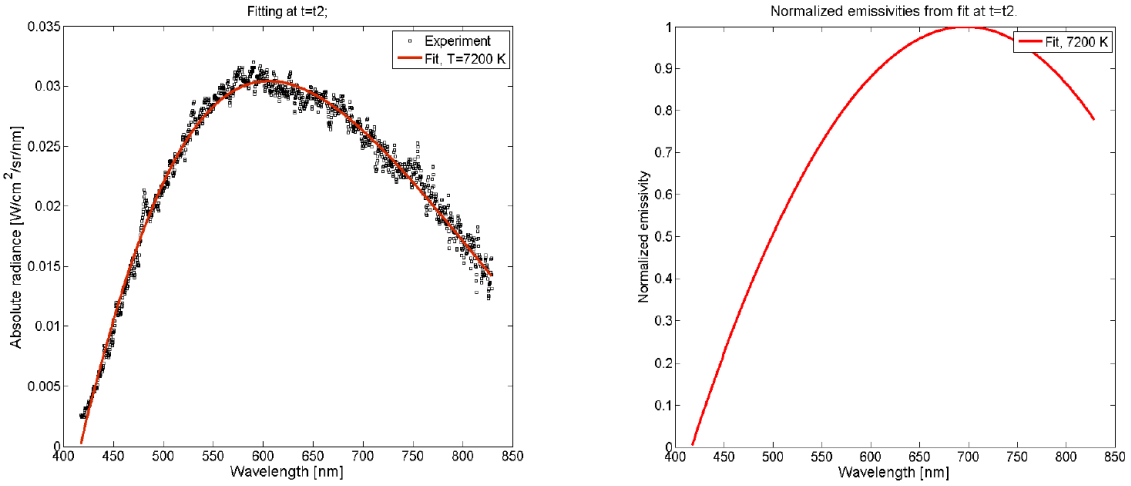

Figure 1: (a) Best fit (with a fitted temperature T = 7200 K) of the spectrometer data(from the data in Fig. 1(b)) using an emissivity assumed to have a quadratic form. **(b)** Inferred best-fit emissivity as function of wavelength.

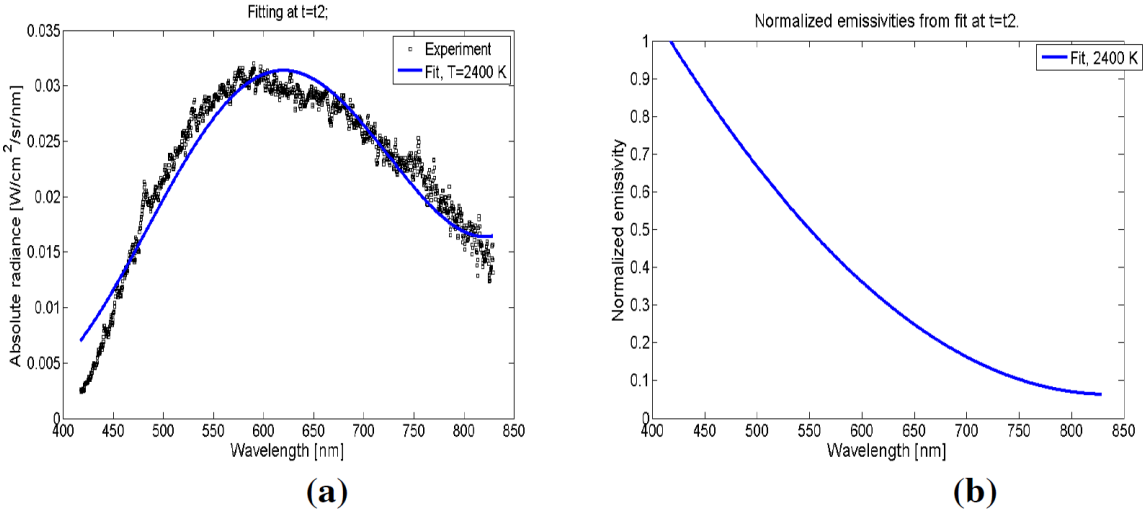

Figure 2: (a) Second "best" fit of the spectrometer data (with a fitted temperature T = 2400 K) (from the data in Fig. 1(b)) using an emissivity assumed to have a quadratic form. **(b)** Inferred fit emissivity as function of wavelength. The parameters derived in figure are also at a minimum in the mean square deviation, but have a larger deviation (worse fit) than those in figure 2.

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